Periodic orbit resonance in (TMTSF)$_2$ClO$_4$

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We have studied the magnetoelectrodynamics of the layered molecular conductor (TMTSF)$_2$ClO$_4$ at millimeter wave frequencies. The high-quality data were obtained in a split-pair magnet using a cavity perturbation technique. We found clear periodic orbit resonance due to the quasi-one-dimensional Fermi surface in the studied compound. These phenomena were investigated in detail. Using our data, we extracted a value for the Fermi velocity which is about 1.8 \( \times 10^5 \) m/s. © 2003 American Institute of Physics.

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Although discovered quite a long time ago, the low-dimensional organic superconductor (TMTSF)$_2$ClO$_4$ continues to be the subject of extensive investigation. This compound belongs to the family of so-called Bechgaard salts with the common formula (TMTSF)$_2$X, where TMTSF is an abbreviation for tetramethyltetraselenafulvalen and the anion X is ClO$_4$, PF$_6$, ReO$_4$, etc.

The crystal structure of (TMTSF)$_2$ClO$_4$ has a quasi-one-dimensional character. The TMTSF cations are packed in columns along the \( a \) axis. These columns form layers parallel to the \( ab \) plane, with the \( c^* \) axis perpendicular to the layers. The strongest interaction between the TMTSF molecules is within the columns; the interaction between the columns and within the layers is about ten times weaker. The weakest coupling exists between the layers.\(^3\) It is about two orders of magnitude bigger than parallel to the layers. As a result, the conductivity along the columns is about three orders of magnitude less than within the columns. The conductivity within the columns is about ten times weaker. The interaction between the TMTSF molecules is within the layers; the interaction between the columns and within the layers is about ten times weaker. The electronic band structure of this compound may be calculated using a tight binding band approximation. These calculations\(^3\) show that the Fermi surface of this salt consists of a pair of weakly warped sheets, which are perpendicular to the \( a \) axis. Thus, the Fermi surface may be regarded as quasi-one-dimensional (Q1D).

For a long time, (TMTSF)$_2$ClO$_4$ was considered to be a conventional superconductor. It has a transition temperature of about 1.2 K, and a finite gap at low temperatures. However, in the last few years, there have appeared some evidences indicating an unconventional character to the superconductivity. For example, Lee et al.\(^6,7\) have shown that the Pauli paramagnetic limit is exceeded in this compound. Therefore, it is very useful to have more detailed information about the Fermi surface of this conductor. The shape of the Fermi surface is determined by the intermolecular electron transfer energies. Their values could be determined from the optical conductivity data using the Drude model. Unfortunately, the applicability of the Drude model is under the question in this compound.\(^8\) More precise data (for example, a transfer integral in the intermediate conducting direction\(^9\)) can be determined from angular-dependent magnetoresistance.

The measurement of the electrodynamics of charge-transfer salts in high fields is the extension of magnetoresistance measurements to high frequencies. For example, some time ago, a periodic orbit resonance (POR) was predicted\(^10\)-\(^12\) and experimentally confirmed for quasi-two-dimensional organic conductors.\(^13\)-\(^15\) It is a unique property of strongly anisotropic metals. In the presence of a magnetic field, an electron moves along the Q1D Fermi surface perpendicular to the magnetic field. The main component of its velocity is directed perpendicular to the Q1D sheets, which corresponds to a movement along the columns. Due to weak coupling between the columns, the Fermi surface sheets are slightly warped. Such warping may be decomposed into the Fourier harmonics, and each harmonic is characterized by some vector \( \mathbf{R} \) in real space.

While an electron moves along the Fermi surface, its velocity parallel to the Q1D Fermi surface sheets oscillates. That velocity component is determined by the warping. For each harmonic, the corresponding velocity oscillates along the warping vector \( \mathbf{R} \). The frequency of these oscillations is proportional to the magnetic-field component perpendicular to both the warping axis and the highest-conductivity direction. Thus, the POR is similar to the usual cyclotron resonance on closed orbits, i.e., the resonance frequency is proportional to the magnetic field. However, instead of the cyclotron mass, the resonance frequency is determined by the Q1D Fermi velocity. There is an equation for the resonance frequency:\(^10,12,15\)

\[
\frac{\nu}{B_{\text{res}}} = \frac{e V_F R}{h} |\sin(\theta)|,
\]

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where $\theta$ is the angle between the projections of magnetic field and the warping axis on the Q1D Fermi surface sheets, $R_i$ is the projection of the lattice period along the warping axis parallel to the Q1D Fermi surface, $B_{res}$ is the resonance field, $V_F$ is the Fermi velocity, and $\nu$ is the measurement frequency. It is a conductivity component parallel to the Q1D Fermi surface which has a maximum at the resonance. The amplitude of the resonance is proportional to the amplitude of the Fermi surface corrugation. So, for strong corrugation, several resonances may be observed. The linewidth of each resonance is proportional to $1/\tau$, where $\tau$ is the relaxation time.\(^{10-12}\) The direction of the warping axes can be determined from the angular dependence of the resonance field. Because the lattice periods are usually well known from x-ray data, the POR measurements represent the most straightforward determination of the Fermi velocity $V_F$.

Although an observation of a possible POR in this compound was reported previously,\(^{16}\) it was not definitively proven. Thus, our motivation was to check if the observed feature is, in fact, the POR on the Q1D Fermi surface, and to determine the Fermi velocity for this salt.

Microwave measurements were carried out using a Millimeterwave Vector Network Analyzer,\(^{15}\) the details of the measurement technique were described previously.\(^{15}\) The sample, with a trapezoid form and dimensions of about 1 $\times$ 0.1 $\times$ 0.2 mm, was placed in a cylindrical cavity. The reported low-temperature value of the interlayer conductivity is about $10^3 / 10^4$ S m\(^{-1}\).\(^{1,19}\) This gives a value of the skin depth at 70 GHz of about 60/190 $\mu$m. Thus, for our experimental configuration, we expect that the cavity absorption due to the sample should be mainly due to interlayer currents. However, we show later that our data suggest otherwise.

The cavity was mounted on the probe, which was placed in a transverse 7 T magnet (i.e., the probe axis was parallel to the field direction). We could smoothly change the sample orientation with respect to the field by \textit{in situ} by rotation of the probe. The rotation plane was either perpendicular to the $a$ axis, or parallel to the $ac^*$ plane. The absolute angle resolution was about 3°. The sample was slowly cooled through the anion ordering transition at 24 K to obtain the relaxed state. The temperature of the measurement was about 2.2 K. Thus, our data were obtained below the so-called field-induced spin density wave transition, in the normal metal regime.\(^{16,20}\)

As shown in Fig. 1, we see one peak in the cavity absorption. Here we plot several traces of the field-dependent absorption, for different sample orientations. The resonance amplitude is almost angular independent, as we can expect from the POR theory.\(^{10-12,15}\) A relative linewidth is roughly constant, which corresponds to the same relaxation time for different angles: $\Delta B / B_{res} \approx 1 / \nu \tau$. We estimate the relaxation time of about 4 ps. This is a typical value for organic metals at low temperatures. Moreover, it is very close to the value reported from the angular dependent magnetoresistance\(^9\) and rapid oscillation\(^{22}\) data.

The angle dependence of the peak position is well described by Eq. (1) for both rotation planes, as shown in Fig. 2.
warping vector has a $2b$ component, the Fermi velocity would be two times smaller.

The origin of the warping vector is unclear at the moment. It may be due to anion ordering, which leads to a complicated warping of the Fermi surface, so that the effective warping vector may not be exactly commensurate with the lattice. More studies of this phenomena will be necessary.

In conclusion, we have demonstrated the presence of a POR in the Q1D organic superconductor (TMTSF)$_2$ClO$_4$ within the normal metallic state. The angle between the warping axis and normal to the layers ($c^*$) is about 66°. The estimated relaxation time is about $4 \times 10^{-12}$ s. Having made the assumption about the warping vector, we determine a value for the Fermi velocity of about $1.8 \times 10^5$ m/s.

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